The Effect of Additional Graphite on the Physical Properties of a Copper Hybrid Compound Using Powder Technique

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ABSTRACT

In this study, copper-3%WC graphite hybrid compounds were produced by powder metallurgy. The effect of different concentrations of graphite (0, 2%, 4%, 6%, 8%) on sinter density and thermal conductivity of the compounds was studied. The results indicated that the sintering density decreased with increasing graphite concentrations by 16.75%, and the thermal conductivity decreased at a rate of 25.90%. This decrease in thermal conductivity is due to the low thermal conductivity of both WC and graphite.

1. Introduction

Due to its high thermal and electrical conductivity, copper and its compounds have found wide applications in the electrical industry, such as high voltage switches, welding electrodes, rocket nozzle liners and kinetic circuit breakers. (Stobrawa et al 2009, Zhao et al 2016, Kovacik et al 2015).

These applications require specific features, such as good mechanical properties and excellent thermal and electrical conductivity; therefore, solid ceramic particles are added to strengthen the copper base (Abu-qaiq et al 2019). Metal overlays such as copper particles and reinforced graphite are candidates for thermal management applications due to their acceptable thermal and electrical conductivity, low thermal expansion coefficient, low density and good machine operation (Unenot et al 2009, Praetor et al 2008).

The high interfacial energy of the Cu graphite system removes moisture between graphite and copper, resulting in phase and pore separation during sintering at high temperatures. To avoid this phenomenon, copper complexes supported with strong carbide elements were used to produce graphite copper complexes. However, adding carbides reduces the thermal conductivity of copper as the thermal conductivity decreases from 400W/m-K to 175 W/m-K (Neubauer 2003, Oku 2007, Veille`re et al 2011).

Copper fittings, which contain oxide and carbide particles, are usually made using powder mining technology. Since this technology is being increasingly used for a number of reasons, including low cost and short processing time, the powders are mixed with a predetermined volumetric fraction, then the powders are combined with cold pressure and sintered at an appropriate temperature, as the sintering temperature depends on the melting temperature Basic Article (Hashemi et al 2016, Stobrawa et al 2009). This work has focused on studying the effect of increasing concentrations of graphite on the thermal conductivity of hybrid copper-based compounds, due to the lack of systematic studies on the thermal conductivity of these compounds as indicated by the researchers, (Kovacik et al 2015) where most studies are interested in studying electrical and mechanical properties (Mehar et al 2017). The present work aims to verify the effect of the graphite content on the sintering density and thermal conductivity of the hybrid compounds produced by powder metallurgy.

2. Materials and Methods

2.1. The Materials Used:

In this study, a copper powder of 99.7 purity and a granular size of 53 µm has been used as the base material for compounds. The first phase of the stiffening particles consists of tungsten carbide with a fixed volumetric fraction and 99.9 purity and granular size 63µm. The second stage of stiffeners consists of variable stone parts of graphite in various proportions (8%, 6%, 4%, 2%, 0) and 99.9 purity and granular size 63 µm.

2.2. Composites Preparation:

The proportions were mixed mechanically for ten minutes and then cold pressed in a steel mould with a diameter of 10mm and one direction by pressing (760 MPa) for a period of half a minute to avoid the occurrence of a flexible return (Lipowsky et al 2007, Habib 2008). After pressing, sintering was performed at 930° C for two hours. The researchers found (Yusoff et al 2013) that the suitable sintering temperature for copper sintering materials supported by tungsten carbide ranges from 850° C to 950° C for two hours.

To prevent oxidation of the samples during flocculation, they were placed in a special ceramic vessel, where a layer of cast iron sculptor 1cm thick was placed on top of it with a layer of silica powder 1cm thick. The oven was turned off and the samples inside the oven were cooled slowly to room temperature (Arheem 2017).

XRD element analysis was examined to find the formed substances, and the densities were measured using the Archimedes method. The thermal conductivity of the compounds was calculated using the Lee’s disc method, and heat was transferred from the heater to the next disc until it reached the last disc. The temperature of the three discs (TA, TB, TC) can be measured by using thermometers inside.
Then, the value of the thermal conductivity coefficient (K) of the sample as a disk is obtained by the following equation (Abdullah et al 2018):

\[
K \left( \frac{T_B - T_A}{d_x} \right) = e \left[ T_A + \frac{2}{r_1} \left( d_A + \frac{1}{4} d_x \right) T_A + \frac{r_1}{\pi} \times d_x \times T_B \right] \quad \ldots \quad \ldots \quad \ldots (1)
\]

Where (e) represents the amount of thermal energy passing through the unit of disk space per second \((w/m^2 {\circ}C)\). It is calculated by the following relationship:

\[
IV = \pi \times r^2 (T_A - T_B) + 2 \pi \times r \times e \left[ d_A \times T_A + d_x \times \frac{T_A + T_B}{2} + d_B \times T_A + d_C \times T_C \right]
\] (2)

Where:
- \((T_A, T_B, T_C)\): The disk temperature (A, B, C), respectively (C)
- \((d)\): thickness of the disc (m)
- \((r)\): radius (m)
- \((I)\): passing current (A)
- \((V)\): fitted voltages (V)

3. Results and Discussion

3.1. X-ray Diffraction:

This looks at XRD analysis of copper-based compounds with fixed volumetric fractions of tungsten carbide and different volumetric fractions of graphite. Figure 1 shows the spectra of X-ray diffraction analysis. The analysis shows strong copper peaks and weak peaks of tungsten carbide and graphite, in addition to weak peaks of copper oxide resulting from the reaction of aerial oxygen trapped in compounds during the sintering process. It can be inferred from the XRD spectra that no interaction occurs between the components, as the base material remains free of any other compounds, which maintains its high thermal conductivity and original mechanical strength.

![XRD Patterns](image)

3.2. Density of Sintering:

Fig (2) shows that the theoretical density and sintering density changed with increasing volumetric fracture content of graphite, where the theoretical density amount decreased by 29% with increasing graphite content, and sintering density decreased by 16.75% when the graphite content increased from 0 to 8%.

3.3. Thermal Conductivity:

It is noted that the first sample that does not contain graphite is higher in density than the rest of the samples, and the sintering density has limits of 8.64g/cm\(^3\). The possible reason for this is the sintering temperature, which reduced the porosity and caused a high density structure consistent with Yusoff et al (2013), where the measured density value at sinter temperature 950ºC was 8.2g/cm\(^3\).

As adding graphite causes a decrease in density, which explains this decrease in sintering density, theoretical density is the tendency of graphite to form clusters in a few locations of the base material. This happens with a higher volumetric fraction of graphite, and graphite also increases the porosity of super positions due to its soft nature, which reduces the sintering density of the compounds. Increasing the graphite content from 0 to 8% causes an increase in the distance between the copper powder, because the melting point of the reinforcing material is higher than the melting point of the base material. It should be noted that the areas of agglomeration formed by graphite act as a barrier against the spread of particle boundaries during the sintering process. The decrease in the interfaces surrounding the base material, due to the increase in the graphite content that lowers the interconnection between the surfaces, impedes the spread of particles during the sintering process. As all these reasons lead to low density of super positions, variation in density appears as the graphite content increases, which corresponds to Varol et al (2015), Rajkumar et al (2011) and Charia et al (2013).

![Effect of graphite content on the theoretical density sintering density](image)
When adding graphite by varying amounts (8%, 6%, 4%, 2%), the thermal conductivity values of the compounds decreased from 205w/m. C° to 152.2w/m. C°. This decrease is due to the low thermal conductivity of graphite and tungsten carbide. However, the measured thermal conductivity is variable, not linear, and this variation is due to the manufacturing method of powder metallurgy, which contains a certain concentration of impurities, most of which are copper oxides (Kovacik et al 2015).

The decrease in densities with the increase in graphite content, where the graphite causes the formation of a base material and an increase in porosity due to the soft nature of the graphite, hinders the spread of particle boundaries. In copper-graphite overlays, the main role of phonons is heat transfer from the graphite phase across the interfaces with the copper base and, when the properties of graphite vary, different types of interfaces can occur. During the P/M manufacturing process, conglomerates of graphite powder are formed, where the graphite powders are congested in a complex manner. Likewise, the interstitial air gaps formed in the super positions directly affect the values of thermal conductivity. Before adding the graphite, the thermal conductivity was high, and this was due to the formation of better interfaces. Densities and interconnect interfaces for super positions (Chu et al 2013).

When the densities decrease, the porosity increases, mainly in the form of pores or separation between the interfaces. This negatively affects the thermal conductivity. Another possible reason is the long sintering time, which affects the thermal conductivity due to the formation of oxides in the compounds. Previous studies (Raza et al 2014, Celebi Efe et al 2011, Vincent et al 2012) indicate that there are scattered values for the measured thermal conductivity.

Finally, the overall causes touched upon include low densities and weak interconnection between the remaining surfaces and (formed) porosity, which is unavoidable in the traditional pressure and sintering process, caused by an increase in the graphite content, as these pores work as insulators inside the superposed body and any increase in their ratio. It works to reduce the thermal conductivity because the pores work to convert the heat transfer by conduction to the heat transfer by the load, which leads to a decrease in the thermal conductivity of the compound.

4. Conclusion

- XRD analysis of the spectra showed strong peaks of copper, in addition to weak peaks of tungsten carbide, graphite and copper oxide formed due to the manufacturing method, and did not show the occurrence of interactions between the base material and the strengthening elements.

- The density test showed that the theoretical density decreased with the increase in the graphite content, and the sintering density decreased by 6.75%.

- The thermal conductivity decreased from 205w/m. C° copper with 3% tungsten carbide to 152w/m. C° when adding graphite. The decrease in thermal conductivity with increasing graphite content was within 25.90%.

Bios

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I obtained a bachelor’s degree in physics in 2000 and a master’s degree in 2005 in the specialty of solid-state physics. I completed my Ph.D. in 2013, also specialising in solid-state physics. Now I am teaching at Tikrit University, College of Education for Pure Sciences, in the Department of Physics. Masters and PhDs inside and outside my college from universities, in addition to my participation in many international and local conferences.

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I obtained a bachelor’s degree from Tikrit University, College of Education, Department of Physics in 1997, and a master’s degree in solid state physics with an excellent grade in 2018 from Tikrit University, College of Education for Pure Sciences, Department of Physics. I work as a teacher of physics in the Salahaddin Education Directorate and worked as a lecturer of laser material at Tikrit University, College of Education Tuz Khurmatu, Department of Physics holds many books of thanks from the Ministry of Education and the Presidency of Tikrit University.

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I obtained a bachelor’s degree from Tikrit University, College of Education, Physics Department in 2013 with a very good grade and completed a master’s degree in solid state physics in 2018 at the same university, also with a very good grade. I work as a teacher of physics in the Kirkuk Education Directorate. I previously worked as a lecturer in electronics and electrical at the College of Education Tuz Khurmatu, Department of Physics, University of Tikrit and received many thanks and appreciation books from the Ministry of Education and the presidency of Tikrit University.

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